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# Efficient, color-stable fluorescent white organic light-emitting diodes with an effective exciton-confining device architecture

Jwo-Huei Jou<sup>\*</sup>, Yung-Sheng Chiu<sup>\*</sup>, Ren-Yang Wang, Huei-Ching Hu,  
Chung-Pei Wang, Hung-Wei Lin

*Department of Materials Science and Engineering, National Tsing Hua University, 101, Section 2 Kuang Fu Road,  
Hsin-Chu 30013, Taiwan, Republic of China*

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## Abstract

Efficient, chromaticity-stable fluorescent white, including pure white, organic light-emitting diodes are fabricated by having an effective exciton-confining device architecture with a single emission layer via vapor-deposition of the solution-mixed targets of a blue light-emitting host doped with a trace amount of a red and/or a green dye. The used host is 1-butyl-9,10-naphthalene-anthracene. The red and green light-emitting dyes are 4-(dicyanomethylene)-2-methyl-6-(julolidin-4-yl-vinyl)-4H-pyran, and 2,3,6,7-tetrahydro-1,1,7,7-tetramethyl-1H,5H,11H-10-(2-benzothiazolyl)quinolizino[9,9a,1gh]coumarin, respectively. The employed electron-blocking effective hole-transporting material is *N,N'*-bis-(1-naphthyl)-*N,N'* diphenyl-1,1'-biphenyl-4-4'-diamine, and the hole-blocking effective electron-transporting material is 2-2'-2''-(1,3,5-benzinetriyl)tris(1-phenyl-1-H-benzimidazole). The best power efficiency of the resulting two-spectrum fluorescent devices is 7.5 lm/W (10.8 cd/A) at 11 cd/m<sup>2</sup> with Commission Internationale de l'Eclairage coordinates of (0.423, 0.426), or 6.5 lm/W (9.6 cd/A) at 12 cd/m<sup>2</sup> with pure white light with coordinates of (0.346, 0.343). The best power efficiency of the three-spectrum counterparts is 6.7 lm/W (9.9 cd/A) at 15 cd/m<sup>2</sup> with a nearly pure white light with coordinates of (0.325, 0.374). All color variations are less than (0.007, 0.006) between 100 cd/m<sup>2</sup> to 10,000 cd/m<sup>2</sup>. © 2005 Elsevier B.V. All rights reserved.

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## 1. Introduction

White organic light-emitting diodes (WOLEDs) have attracted much interest for having great potential for use in high-quality flat-panel displays, plane illumination, liquid-crystal-display backlighting and opto-electronic applications [1–4]. The power

<sup>\*</sup> Corresponding authors. Tel.: +886 3 5719524; fax: +886 3 5742617.

E-mail addresses: [jjou@mx.nthu.edu.tw](mailto:jjou@mx.nthu.edu.tw) (J.-H. Jou), [d907512@oz.nthu.edu.tw](mailto:d907512@oz.nthu.edu.tw) (Y.-S. Chiu).

efficiency and lifetime of WOLEDs are currently the primary concerns [5–7]. The emitting layer can be made of phosphorescent [8–10] and/or fluorescent [11–16] materials. Phosphorescent WOLEDs may be suitable for the future illumination for having shown very high luminance and power efficiency [4]. The best reported power efficiency of phosphorescent WOLEDs is 26 lm/W [10]. The best reported power efficiency, however, is 5.0 lm/W for three-spectrum fluorescent WOLEDs with pure white emission [11]. The efficiency is 5.35 lm/W with pure white emission, while 6.0 lm/W with a near pure white emission for two-spectrum fluorescent WOLEDs [12,13]. The power efficiency of fluorescent WOLEDs is comparatively low. Fluorescent WOLEDs with a much-improved efficiency are thus being pursued.

To increase luminous efficiency, the host must first exhibit efficient energy-transfer capability in triggering the emissions of the various dyes [17]. Second, the host is better coupled with appropriate electron- and hole-transporting materials to form low-energy barriers, thus enabling electrons and holes, respectively, to enter the emission layer easily. Third, electron- and hole-blocking functions are provided therein to effectively confine more electrons and holes having entered the desired emission zone to further increase the electron-hole recombination probability, and the corresponding efficiency [18–20].

This work reports the fabrication of efficient, color-stable, white and pure white fluorescent organic light-emitting diodes with an effective exciton-confining device architecture with a single emission layer by vapor-deposition of solution-mixed deposition targets. The best power efficiency of the resulting two-spectrum fluorescent devices is 7.5 lm/W (10.8 cd/A) at 11 cd/m<sup>2</sup> with Commission Internationale de l'Eclairage (CIE) coordinates of (0.423, 0.426), or 6.5 lm/W (9.6 cd/A) at 12 cd/m<sup>2</sup> with a pure white light of (0.346, 0.343). That of the three-spectrum counterparts is 6.7 lm/W (9.9 cd/A) at 15 cd/m<sup>2</sup> with a nearly pure white light of (0.325, 0.374). All color variations are less than (0.007, 0.006) between 100 cd/m<sup>2</sup> to 10,000 cd/m<sup>2</sup>.

## 2. Experimental

Fig. 1 depicts the WOLED structure studied and the organic molecules used. The devices comprise a 125 nm indium tin oxide (ITO) anode, a 45 nm hole-transporting layer (HTL) of *N,N'*-bis-(1-naphthyl)-*N,N'* diphenyl-1,1'-biphenyl-4-4'-diamine (NPB), a 30 nm white emission layer, a 40 nm electron-

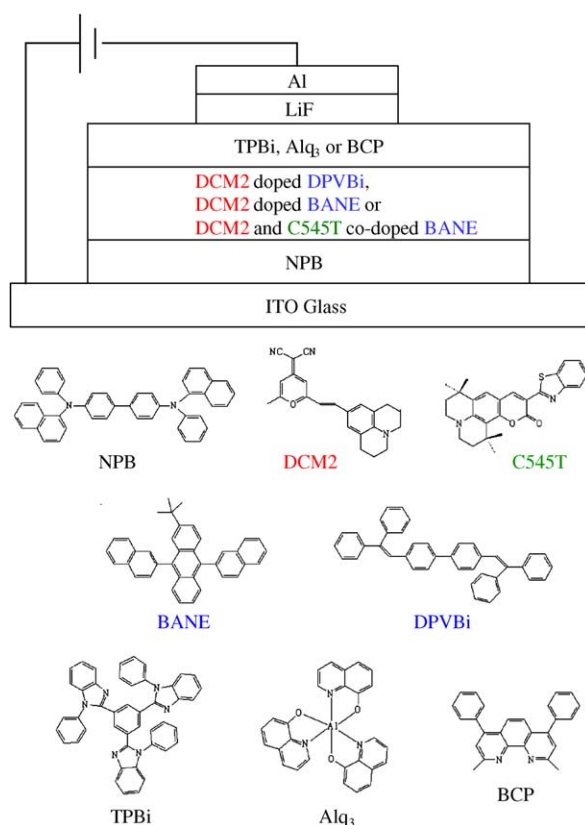


Fig. 1. Schematic illustrations of the structures of the two- or three-wavelength fluorescent WOLEDs and the organic molecules studied.

transporting layer (ETL) of 2-2'-2''-(1,3,5-benzinetriyl)tris(1-phenyl-1-H-benzimidazole) (TPBi), a 0.5 nm electron-injecting layer of lithium fluoride and a 150 nm aluminum cathode. The white emission layer is formed by vapor-deposition of the solution-mixed deposition target composed of the blue host 1-butyl-9,10-naphthalene-anthracene (BANE) with a trace amount of red dye 4-(dicyanomethylene)-2-methyl-6-(julolidin-4-yl-vinyl)-4H-pyran (DCM2). To form three-spectrum white emission, an additional trace amount of green dye 2,3,6,7-tetrahydro-1,1,7,7-tetramethyl-1H,5H,11H-10-(2-benzothiazolyl)quinolizino[9,9a,1gh]coumarin (C545T) is added. Another blue host of 1,4-bis(2,2-diphenylvinyl)biphenyl (DPVBi) and two other ETLs of bathocuproine (BCP) and tris(8-hydroxyl-quinoline)aluminum (Alq<sub>3</sub>) are studied for comparison.

The deposition source of the white emission layer of two- or three-spectrum was prepared via solution-mixing [21] as followed. The composing dye(s) and host were first separately dissolved in an

organic solvent such as tetrahydrofuran or toluene. After complete dissolution, the resultant solutions were then mixed to form a host solution uniformly dispersed with the desired doping dye(s). Low doping concentrations were obtained by using the diluted dye solutions. The resulted dye-dispersed host solution was then vacuum-dried at 80 °C for 60 min prior to vapor-deposition.

The devices were fabricated by vapor-deposition using an ITO coated glass substrate (Merck Display Technologies, Ltd.) with a sheet resistance of 13  $\Omega$ /square. The substrate was cleaned in ultrasonic baths of detergent, de-ionized water, acetone and isopropyl alcohol in turn, and then treated with the boiling hydrogen peroxide. The substrate was then dried by purging nitrogen. The respective organic layers and the cathode layer were deposited at  $2 \times 10^{-5}$  Torr using resistively heated tantalum and tungsten boats. All the organic layers were deposited at rates ranging from 0.1 to 0.3 nm/s. The lithium fluoride and aluminum cathode were subsequently deposited at rates of 0.01 and 1 nm/s, respectively. The emission area of all the devices was 8 mm<sup>2</sup>, and only the luminance in the forward direction was measured.

The luminance and Commission International de L'Eclairage (CIE) chromatic coordinates of the resulted OLEDs were measured by using Minolta CS-100 luminance-meter. The electroluminescence and photoluminescence spectra were measured using a Hitachi F-4500 fluorescence spectrophotometer. The ultraviolet visible (UV–VIS) absorption spectra of the DCM2 and C545T solutions were measured using a Hitachi U-3010 UV–VIS spectrophotometer. The highest occupied molecular orbital (HOMO) energy levels of the organic materials studied were calculated from their oxidation potentials measured by a cyclic voltammetry [22], while the corresponding lowest unoccupied molecular orbital (LUMO) energy levels were estimated based on their HOMO energy levels and the lowest-energy absorption edge of the UV–VIS absorption spectra. A Keithley 2400 electrometer was used to measure the current-voltage ( $I$ – $V$ ) characteristics. All the devices were characterized without packaging and all the measurements were carried out in the ambient condition.

### 3. Results and discussion

Fig. 2 shows the effect of the DCM2 concentration on the resultant electroluminescence spectra.

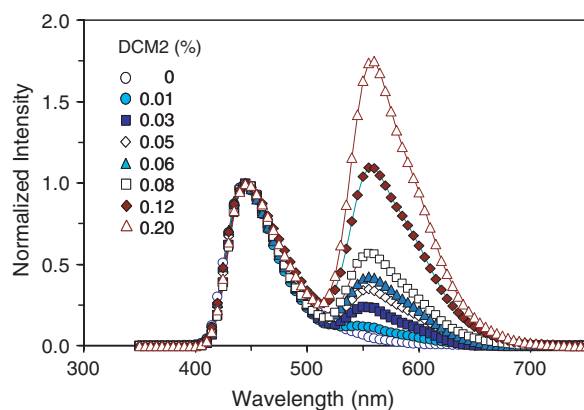


Fig. 2. Doping-concentration effects of the 'red' dye of DCM2 on the electro-luminescence spectra of the two-spectrum WOLEDs.

The spectra exhibit two major emission peaks at 450 nm from the blue host, BANE, and at 560 nm from the red-dopant, DCM2, respectively. The emission that yields pure DCM2 (100%) peaks at 630 nm is strongly blue-shifted when its doping-concentration becomes extremely low, ranging from 100 to 2000 ppm, which is due to a strong solid state solvation effect [23]. The marked blue-shift turned the DCM2, itself, from a so-called red dye to an orange one in these doping ranges.

Table 1 presents the effects of the concentration of the various light-emitting dopants on the resultant luminous characteristics. The resultant CIE coordinates change from that of blue light (0.155, 0.122) to that of yellow light (0.464, 0.457) as the DCM2 concentration increases from 0 to 2000 ppm. The changing path passes through the pure white light region,  $(0.33 \pm 0.03, 0.33 \pm 0.03)$ , indicating that pure white light can be obtained from these device systems if they are of proper composition. For example, the lower and upper limits are 300 and 1200 ppm, respectively, to emit white light, while only 500–600 ppm of red-dopant DCM2 can be presented to emit pure white light with emission between (0.330, 0.322) and (0.346, 0.343).

The maximum power efficiency, which is defined as the value obtained at a luminance of  $\geq 10$  cd/m<sup>2</sup>, increases from 1.3 to 8.3 lm/W as the DCM2 concentration increases from 0 to 2000 ppm for devices A to H. Pure white emission of (0.346, 0.343) with a power efficiency of 6.5 lm/W (9.6 cd/A) at 12 cd/m<sup>2</sup> is obtained by doping with 600 ppm of DCM2 (device E). A much higher power efficiency, 7.5 lm/W (10.8 cd/A) at 11 cd/m<sup>2</sup>, is obtained by

Table 1

The effects of doping-concentration of the various light-emitting dopants on the luminous characteristics of the fluorescent WOLEDs having a single emission layer composing of the blue host of BANE doped with trace amounts of the 'red' dye of DCM2 and/or the green dye of C545T

No.	DCM2 (%)	C545T (%)	Max. luminance (cd/m <sup>2</sup> )	Maximum power/ current efficiency <sup>a</sup>		CIE1931 (x,y) chromatic coordinates at	
				(lm/W)	(cd/A)	100 cd/m <sup>2</sup>	10,000 cd/m <sup>2</sup>
A	–	–	6500	1.3	3.6	(0.155,0.122)	–
B	0.01	–	9900	2.1	3.3	(0.192,0.162)	(0.198,0.169)
C	0.03	–	15,800	5.0	7.3	(0.280,0.269)	(0.276,0.263)
D	0.05	–	16,900	5.5	7.6	(0.330,0.322)	(0.328,0.317)
E	0.06	–	19,600	6.5	9.6	(0.346,0.343)	(0.341,0.337)
F	0.08	–	24,700	6.5	9.7	(0.382,0.381)	(0.380,0.380)
G	0.12	–	39,500	7.5	10.8	(0.423,0.426)	(0.418,0.420)
H	0.20	–	41,500	8.3	13.4	(0.464,0.457)	(0.455,0.452)
I	0.50	–	39,300	7.6	12.1	(0.479,0.471)	(0.469,0.466)
J	1.00	–	38,900	7.3	11.6	(0.485,0.472)	(0.477,0.469)
K	2.00	–	29,500	7.2	11.4	(0.505,0.467)	(0.498,0.467)
L	0.05	0.02	23,200	5.7	8.1	(0.320,0.347)	(0.316,0.342)
M	0.05	0.04	24,200	6.5	9.3	(0.325,0.374)	(0.320,0.370)
N	0.05	0.06	25,300	6.7	9.9	(0.324,0.395)	(0.317,0.389)
O	0.05	0.08	23,000	6.9	10.7	(0.316,0.411)	(0.305,0.401)
P	0.05	0.12	22,500	7.2	11.3	(0.321,0.439)	(0.312,0.432)

<sup>a</sup> The maximum power/current efficiency is defined as the value obtained at a luminance of  $\geq 10$  cd/m<sup>2</sup>.

raising DCM2 to 1200 ppm (device G). However, the corresponding color is not pure white, but is within the generally defined white emission region with CIE coordinates of (0.423,0.426). These two devices exhibit the highest power efficiency of all reported two-spectrum fluorescent WOLEDs.

The power efficiency, however, decreases from 8.3 to 7.2 lm/W as the DCM2 concentration increases from 0.2% to 2% for devices H to K. This decreasing phenomenon may be attributed to the increasing formation of guest molecule aggregates [23] and low luminescent-efficiency excimers [24] at higher doping-concentrations. The combined effect of these two mechanisms would result in a lower luminous efficiency, which is typically known as concentration-quenching [25]. Moreover, carrier trapping frequently exists in the EL process of dye doped systems [26,27]. It would be more likely to occur when the doping-concentration is higher. This mechanism may also be taken into account for the aforementioned efficiency-decreasing phenomenon.

The chromaticity of emission from all resulting two-spectrum WOLEDs (devices C, D, E, F and G) changes slightly between 100 and 10,000 cd/m<sup>2</sup>. The emission coordinates of the device E, for example, are (0.346,0.343) at 100 cd/m<sup>2</sup> and (0.341,0.337) at 10,000 cd/cm<sup>2</sup>, respectively. The chromatic variation is as small as (0.005,0.006), which is the least ever reported for pure white OLEDs. The

chromaticity of all other devices varied less. Of these, device F, which contains 800 ppm DCM2 emits white light (0.382,0.381), exhibits the least variation of (0.002,0.001).

Three reasons may explain the relatively high color-stability of the WOLEDs. Importantly, the devices simply compose one emission layer [12,15,16]. The entering electrons/holes can be effectively confined and so recombine within the desired emission layer owing to the presence of two neighboring electron-blocking and hole-blocking layers. The recombination zone will hence barely shift as the applied voltage increases. Second, the guest molecules are uniformly dispersed throughout the entire thickness in the emission layer by employing the solution-mixing method in the preparation of the vapor-deposited target. A zone shift caused by applied-voltage-variation occurs mostly within the confined emission layer, explaining why the resulting colors are quite the same. Third, all the resulted WOLEDs employed comparatively low doping-concentrations. Less exciton quenching would have occurred and much less color variation was then observed as the current density increased [26].

Fig. 3 shows the effect of C545T concentration on the electroluminescence spectrum of the resultant three-spectrum WOLEDs. The spectra show three emission peaks at 450 nm from the blue host of BANE, at 495 nm from the green dopant of

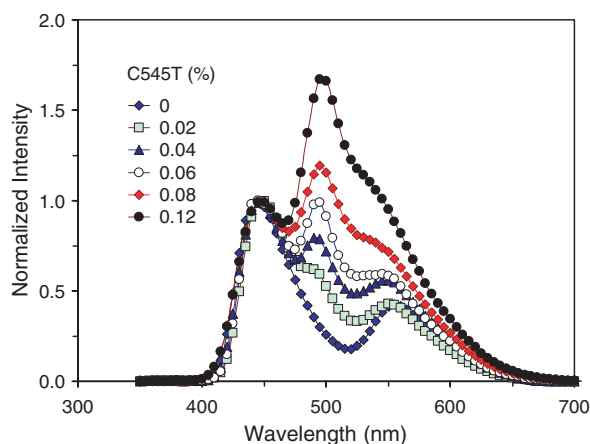


Fig. 3. Doping-concentration effects of the green dye of C545T on the electro-luminescence spectra of the three-spectrum WOLEDs.

C545T and at 560 nm from the red dopant of DCM2. The emission changes from a pure white light of (0.330,0.322) to the upper boundary of white light with an emission of (0.324,0.395) as the C545T concentration is increased from 0 to 600 ppm. Increasing the C545T concentration to 1200 ppm causes the emission to turn to green with chromatic coordinates of (0.321,0.439).

The maximum power efficiency increases from 5.5 lm/W (7.6 cd/A) to 7.2 lm/W (11.3 cd/A) as the C545T concentration is increased from 0 to 1200 ppm. Device L, which contains 200 ppm of C545T, emits pure white light with a maximum power efficiency of 5.7 lm/W (8.1 cd/A) at 13 cd/m<sup>2</sup>. A much higher power efficiency, 6.7 lm/W (9.9 cd/A) at 15 cd/m<sup>2</sup>, is obtained by increasing the doping C545T to 600 ppm (device N). The corresponding emission remains white but no longer pure white. All the resulting three-spectrum WOLEDs exhibit very little change in color between 100 and 10,000 cd/m<sup>2</sup>. For example, for devices L and N, the corresponding chromatic coordinates change from (0.320,0.347) to (0.316,0.342) and from (0.325,0.374) to (0.320,0.370), respectively. The corresponding chromatic variations are (0.004, 0.005) and (0.005, 0.004), respectively. The largest chromatic variation observed herein is (0.007, 0.006), i.e., smaller than those of all previously reported three-spectrum WOLEDs.

White emission can be obtained in both the two- and three-spectrum OLEDs. Taking the two-wavelength OLEDs for examples, the white emission is resulted from the mixing of the red, or more

correctly orange, emission from the red dye and the blue emission from the blue light-emitting host. The concentration of DCM2 is so low that the coming carriers of hole and electron must have mostly gone through and recombined on the host molecules. The excited energy upon recombination has apparently transferred at least partly from the host to the guest of DCM2 to yield the observed orange emission, according to Förster energy transfer mechanism [28]. And, part of the excited energy is used to trigger its own blue emission as evidenced by the blue emission peak at 450 nm shown in Fig. 3. Similarly, the doping-concentration of green dye C545T is also very low and its green emission is triggered by the excited energy transferred from the host molecules via the same mechanism.

Fig. 4 shows the effect of host materials on the resultant luminous characteristics. The power efficiency is 6.7 lm/W at 0.05 mA/cm<sup>2</sup> for the BANE host, while 5.5 lm/W at 0.18 mA/cm<sup>2</sup> for the DPVBi host. Their respective CIE chromatic coordinates at 100 cd/m<sup>2</sup> are (0.330,0.322) and (0.337,0.349). The devices with these two different hosts yield almost, but not exactly, the same pure white emission. The minor difference is attributable to the difference between the colors originally emitted by the hosts, themselves. Pure BANE has chromatic coordinates of (0.155,0.122) and pure DPVBi has ones of (0.170,0.165). BANE is a host with a deeper blue color than DPVBi, so BANE is a more appropriate host for the 'red' dye, DCM2, to generate a pure white light.

Fig. 5 shows the photoluminescence spectra of the host molecules of BANE and DPVBi and the UV–VIS absorption spectra of the guest molecules

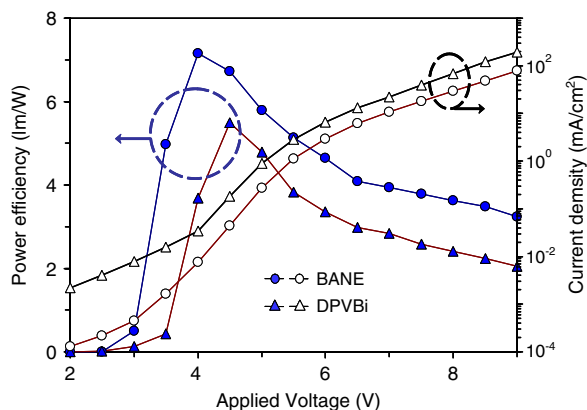


Fig. 4. The effects of the different hosts of BANE and DPVBi on the resulting power efficiency, current density of the WOLEDs using the two different hosts.



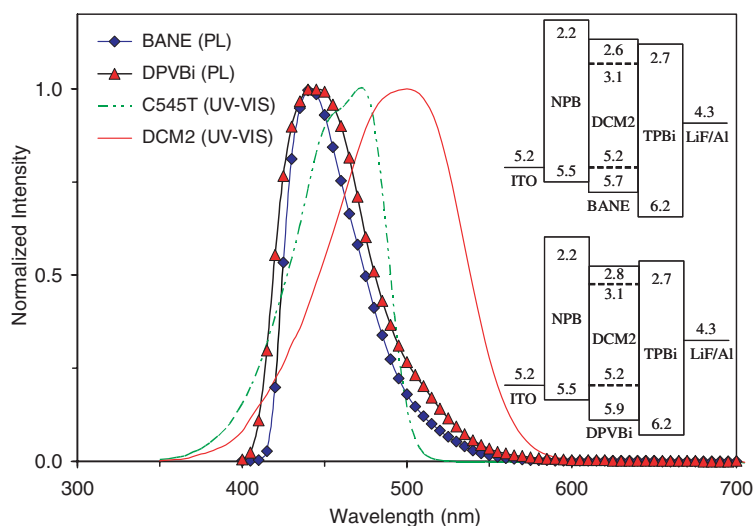


Fig. 5. The photoluminescence spectra of the host molecules of BANE and DPVBi, ultraviolet–visible absorption spectra of the guest molecules of DCM2 and C545T, and HOMO/LUMO energy-level alignments (inset) of the WOLEDs using the two different hosts.

of DCM2 and C545T. The spectral peak from the BANE host ranges from 415 to 580 nm, and that of DPVBi host ranges from 405 to 590 nm. The absorption peak of red-dopant DCM2 ranges from 380 to 585 nm, and that of green-dopant C545T from 370 to 515 nm. The photoluminescence spectra of both hosts overlap greatly with the UV–VIS absorption spectra of the two guest molecules, revealing the likeliness of high-efficiency energy-transfer [17]. Although the overlapping area of the host DPVBi is larger than that of the host BANE, the resulting efficiency is slightly lower than that of the BANE counterpart, revealing that other factors influence the overall efficiency of the device, as described below.

The electrons are initially injected from the cathode into the electron-transporting layer of TPBi, as shown by the energy-level alignment of the resultant device structure in the inset in Fig. 5. The LUMO of TPBi is 2.7 eV, and that of the emitting layer of BANE is 2.6 eV. An energy barrier of 0.1 eV must be overcome to transport the electrons from the TPBi layer into the BANE host. The LUMO of the other host, DPVBi, is 2.8 eV. The energy barrier is  $-0.1$  eV, revealing that no barrier exists for the electrons to enter the host of DPVBi. These facts explain why the exhibited current density is obviously markedly higher for the device employing the DPVBi host (Fig. 4). Although possessing better electron-transporting characteristics, the device with a DPVBi host does not have a correspondingly higher luminous efficiency. This finding suggests

yet another crucial factor affecting the resultant efficiency.

Also shown in the insert of Fig. 5, the holes are initially injected from the anode to the hole-transporting layer of NPB. The HOMO of NPB is 5.5 eV, while that of the BANE host is 5.7 eV. The energy barrier to the transport of holes from the NPB layer into the BANE host is 0.2 eV, whereas the barrier to entering the DPVBi host is 0.4 eV. Clearly, the holes can more easily enter the BANE host. Additionally, the energy barrier to the further transport of holes from the BANE host to the next layer, TPBi (HOMO = 6.2 eV), is 0.5 eV, whereas the barrier to transport from the DPVBi host is 0.3 eV. The higher energy barrier, 0.5 eV, would subsequently make it more difficult for the holes to leave the BANE host. More holes are then confined in the BANE host, increasing the electron-hole recombination probability and the luminous efficiency. These results explain why WOLEDs with a BANE host have a higher luminous efficiency.

Both energy transfer and carrier trapping exist simultaneously in the emission process [26,27]. The occurrence of carrier trapping on the dopants would reduce the total efficiency. Nevertheless, the above WOLEDs that employ BANE and DPVBi hosts both exhibit relatively high efficiency. This may be attributed to the comparatively low doping concentration used for the red dye of DCM2 since the occurrence of carrier trapping would be less [27].

Fig. 6 shows the effect of the electron-transporting material on the luminous characteristics. The

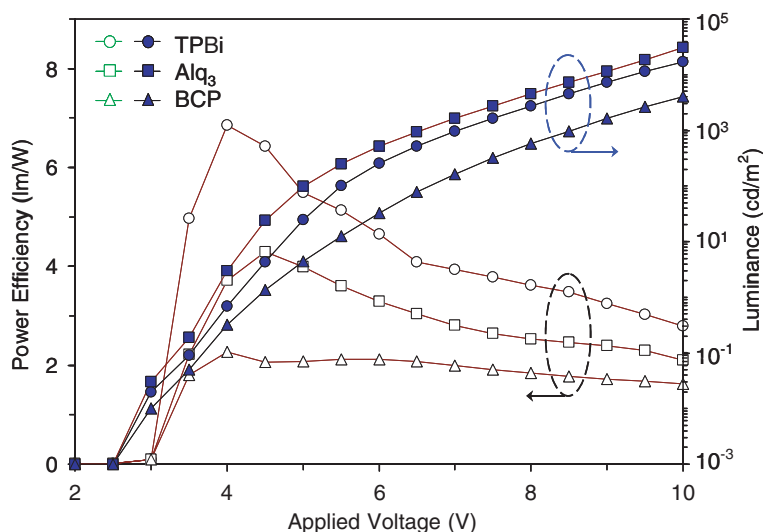


Fig. 6. The effects of the different electron-transporting materials of TPBi, BCP and Alq<sub>3</sub> on the resulting power efficiency and luminance of the two-spectrum WOLEDs.

maximum power efficiencies of WOLEDs with the electron-transporting layers of TPBi, Alq<sub>3</sub> and BCP are 6.8, 4.3 and 2.5 lm/W, respectively. Their corresponding luminances are 16,900, 30,700 and 4000 cd/m<sup>2</sup> at 10 V. The energy barrier to the transport of electrons from TPBi to the same BANE host is 0.1 eV, while the barrier to transport from Alq<sub>3</sub> (LUMO = 3.0 eV) is 0.4 eV and that from BCP is

0.6 eV (LUMO = 3.2 eV) as shown in Fig. 7. Consequently, the devices with a TPBi electron-transporting layer have the highest efficiency for having the lowest electron-injection barrier. Although BCP provides the best hole-blocking function, its huge electron-injection barrier, however, strongly hinders the entry of electrons into the emitting host.

The device using Alq<sub>3</sub> has yielded a poor efficiency for having as well a high electron-injection barrier. Nevertheless, its brightness is the greatest among all these devices. This may be attributed to its poorest hole-blocking effect that the unrecombined holes can easily enter the green light-emitting Alq<sub>3</sub> layer to recombine with the electrons injected from the cathode, substantially increasing the brightness, but changing the color to an undesired green.

#### 4. Conclusion

In conclusion, we have presented the fabrication of efficient, color-stable, two- and three-spectrum white and pure white fluorescent organic light-emitting diodes with an effective exciton-confining device architecture with a single emission layer by vapor-deposition of the solution-mixed deposition targets of an efficient blue host of BANE doped with the red dye of DCM2 and/or green dye of C545T. The best power efficiency of the resulting two-spectrum fluorescent WOLEDs is as high as 7.5 lm/W (10.8 cd/A) at 11 cd/m<sup>2</sup> with chromatic

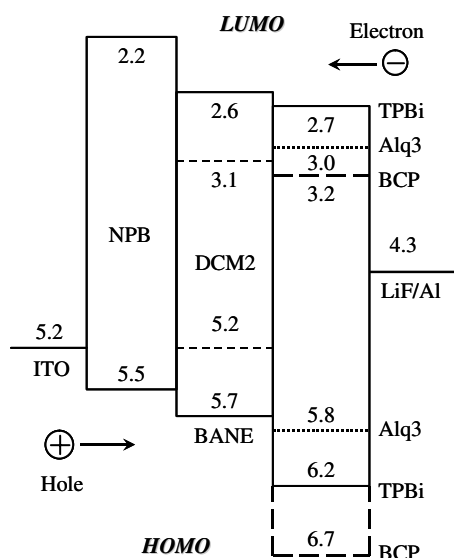


Fig. 7. The HOMO/LUMO energy-level alignment of the two-spectrum WOLEDs with the three different electron-transporting materials of TPBi, BCP and Alq<sub>3</sub>.

coordinates of (0.423, 0.426), or 6.5 lm/W (9.6 cd/A) at 12 cd/m<sup>2</sup> with a pure white light of (0.346, 0.343). The best power efficiency of the three-spectrum counterparts is 6.7 lm/W (9.9 cd/A) at 15 cd/m<sup>2</sup> with a nearly pure white light of (0.325, 0.374). All color variations are less than (0.007, 0.006) between 100 and 10,000 cd/m<sup>2</sup>.

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